Carrier Density Control of Magnetism and Hall Effects in $EuTiO_3$ Films

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Outline

- Spontaneous Hall effects in magnetic materials
- Rare-earth doped $EuTiO_3$ films
  - Fermi surface topology, magnetism, and quantum criticality
- $SmTiO_3$/$EuTiO_3$ interfaces
- Summary

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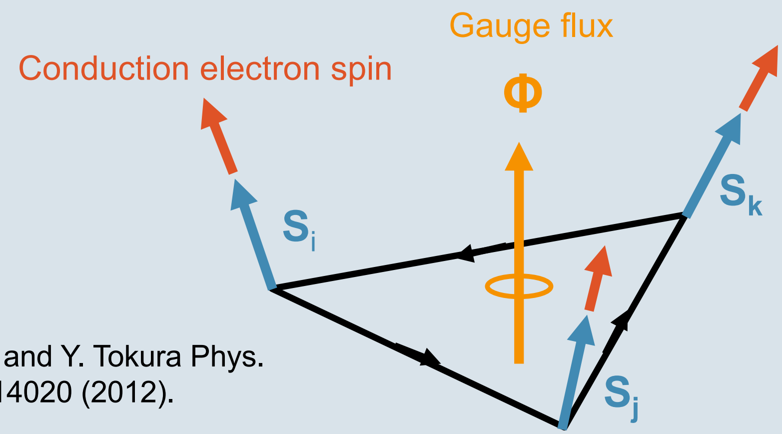
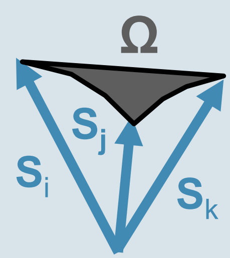
Berry Curvature and Hall Effects

Berry phase associated with a non-coplanar spin structure

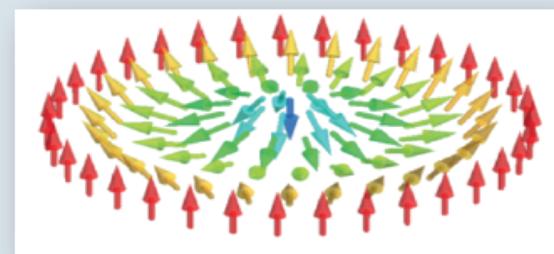


N. Nagaosa and Y. Tokura Phys. Scr. **T146** 014020 (2012).

Spin chirality
$S_i \cdot (S_j \times S_k)$



Skyrmion



- Berry phase of the magnetization texture
- Electron spins of the conduction electrons are coupled adiabatically and therefore accumulate a Berry phase
- Effective Lorentz force results in a “Topological Hall effect” P. Bruno, et al., PRL 93, 096806 (2004).

Topological Hall effect

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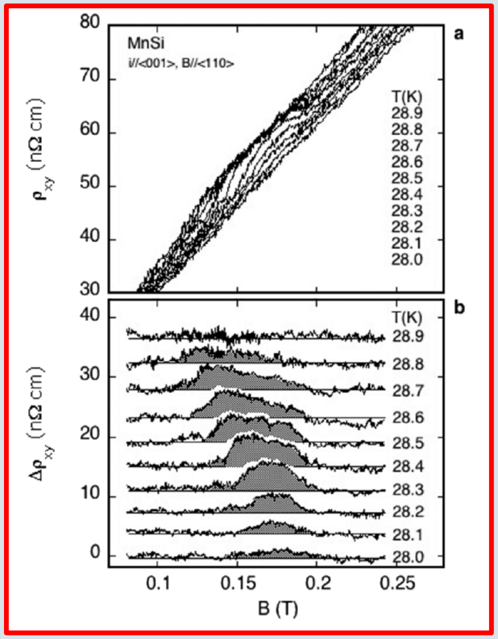
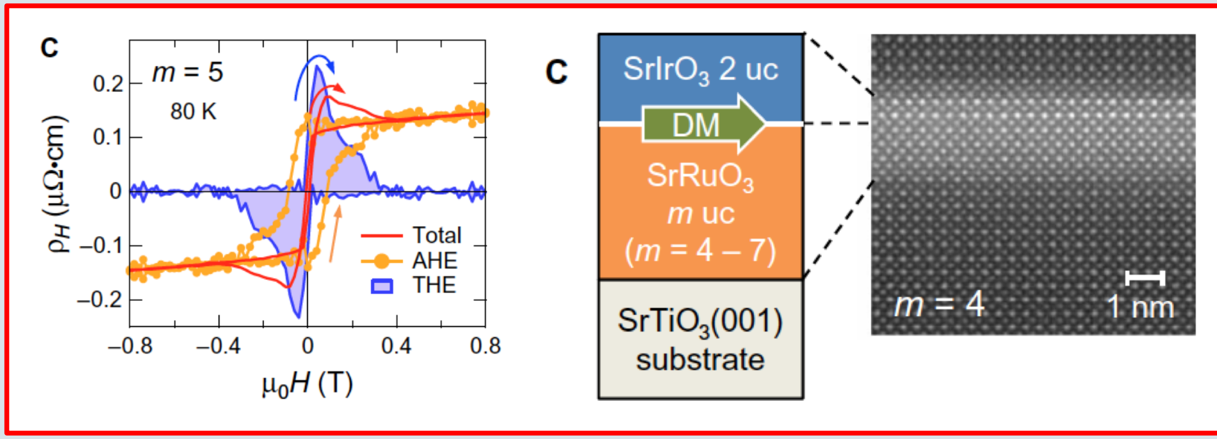
Experimental Observation of the Topological Hall Effect

Theory: Ye et al., PRL 83, 3737 (1999); Y. Taguchi et al., Science 291, 2573 (2001); Onoda et al., PRL 90, 196602 (2003); P. Bruno, et al., PRL 93, 096806 (2004); B. Binz et al., Physica B 403, 1336 (2008).



MnSi - Skyrmion Phase

A. Neubauer, et al., PRL 102, 186602 (2009).



$SrIrO_3$/$SrRuO_3$ Interface Skyrmions

Matsuno et al., Sci. Adv. 2, e1600304 (2016).

Topological Hall effect

Berry Curvature and Hall Effects

Berry phase in reciprocal space

R. Karplus and J. M. Luttinger: Phys. Rev. 95 1154 (1954).
M.-C. Chang and Q. Niu, PRL 75, 1348 (1995).
F. D. M. Haldane, PRL 93, 206602 (2004).
Nagaosa et al., Rev. Mod. Phys. 82, 1539 (2010).

Equation of motion in an external field:

*velocity in state **k***

$$\boldsymbol{v_n}(\mathbf{k}) = \underbrace{\frac{\delta\epsilon_n(\mathbf{k})}{\hbar\delta(\mathbf{k})}}_{\text{Band dispersion}} - \underbrace{\frac{\mathrm{e}}{\hbar}\boldsymbol{E}\times\boxed{\boldsymbol{\Omega}_n(\mathbf{k})}}_{\text{Anomalous velocity}}$$

⇨ *Hall effect*

- $\Omega_n(\mathrm{k})$ is the Berry curvature of the nth band
- Acts in reciprocal space like a magnetic flux density in real space: "reciprocal space magnetic field"
- **$\boldsymbol{\Omega_n}(\mathrm{k})$ is a band structure property, in addition to the usual band dispersion**
- Non-zero in systems with broken time-reversal symmetry (magnetic materials) OR broken space inversion symmetry

Intrinsic Anomalous Hall Effect

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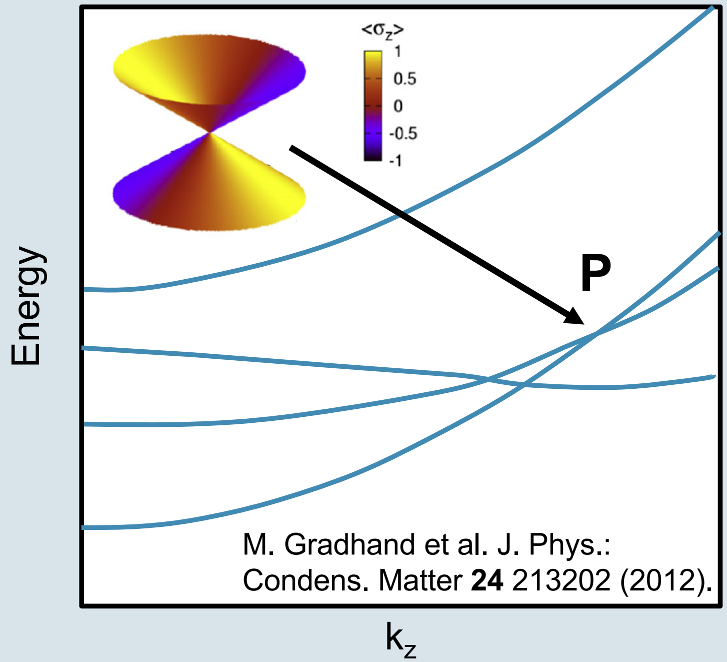
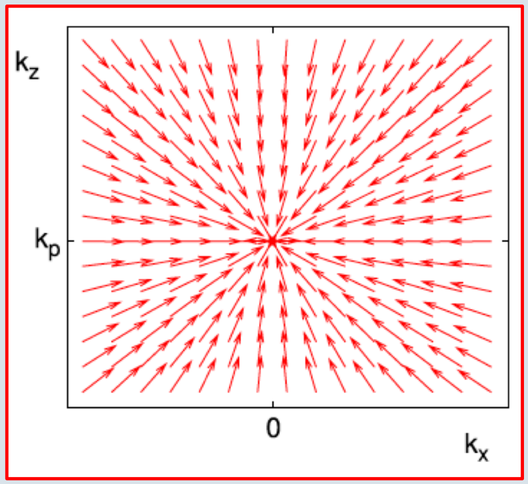
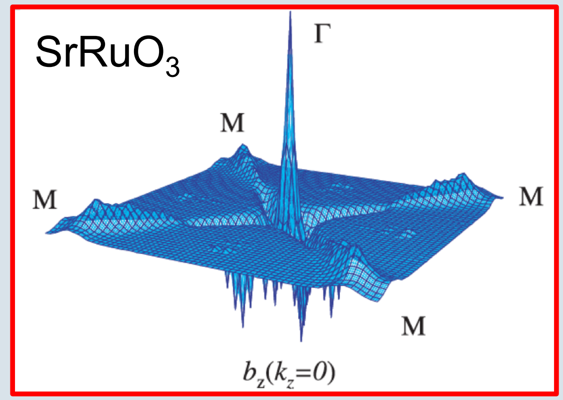
Berry Curvature in Reciprocal Space



M. Gradhand et al. J. Phys.: Condens. Matter **24** 213202 (2012).

Magnetic monopoles in reciprocal space

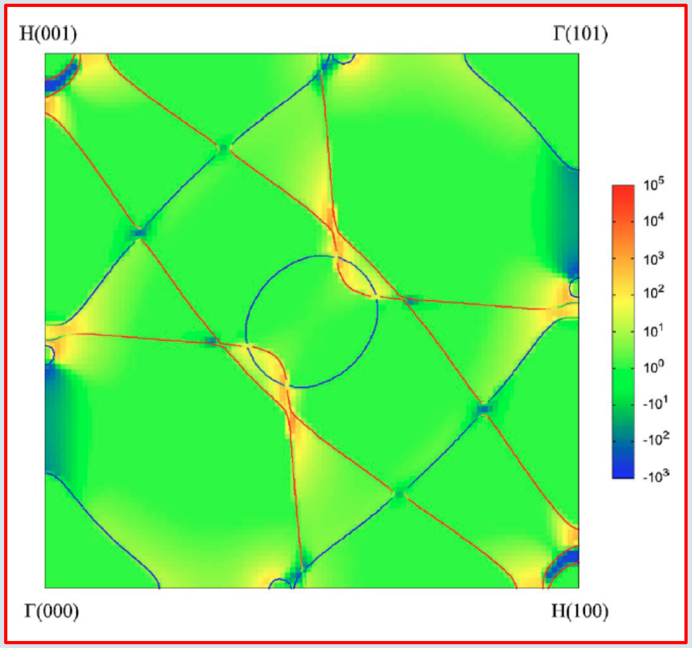




Fang et al., Science 302, 92 (2003).

- Band crossings or avoided band crossings cause large variations in spin with **k** and are sources/sinks of Berry curvature
- Magnetic monopoles in reciprocal space

Berry Curvature in Reciprocal Space



The calculated Berry curvature is shown in Fig. 6. It can be seen that the regions in which the Berry curvature is small (light green regions) fill most of the plane. The largest values occur at the places where two Fermi lines approach one another, consistent with the discussion of Fig. 3. Of special importance are the avoided crossings between two bands having the same sign of spin, or between two bands of opposite spin. Examples of both kinds are visible in the figure, and both tend to give rise to very large contributions in the region of the avoided crossing. Essentially, the spin-orbit interaction causes the character of these bands to change extremely rapidly with **k** near the avoided crossing; this is the origin of the large Berry curvature.

Wang et al., PRB **74**, 195118 (2006).

- Band crossings or avoided band crossings cause large variations in spin with **k** and are sources/sinks of Berry curvature

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Anomalous Hall Effect

$$v_n(\mathbf{k}) = \underbrace{\frac{\delta\epsilon_n(\mathbf{k})}{\hbar\delta(\mathbf{k})}}_{\text{Band dispersion}} - \underbrace{\frac{e}{\hbar}\boldsymbol{E}\times\boxed{\boldsymbol{\Omega}_n(\mathbf{k})}}_{\text{Anomalous velocity}}$$

➡ ***Anomalous Hall effect***

- **Ω_n(k) is a band structure property, in addition to the usual band dispersion**
- **Anomalous Hall effect even in antiferromagnets with small net magnetization**

H. Chen, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. 112, 017205 (2014); J. Kübler and C. Felser, Europhys. Lett. 108, 67001 (2014). S. Nakatsuji , N. Kiyohara and T. Higo, Nature 527, 212 (2015).

- **Expect the intrinsic anomalous Hall effect to depend on Fermi level position**

  Z. Fang et al., Science 302, 92 (2003).

→ **Control via carrier density (doping, electric field effect, …)**

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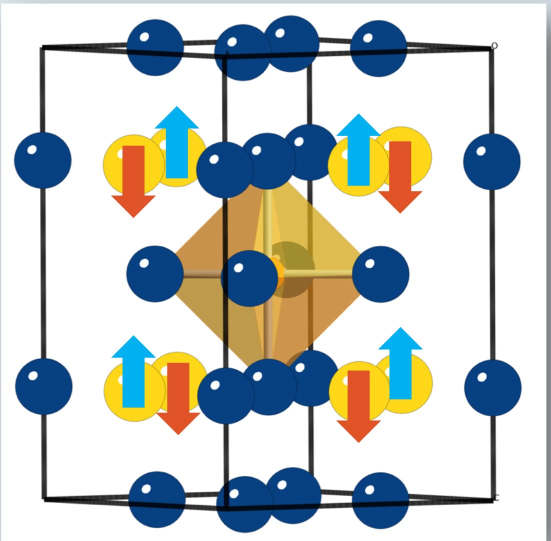
Rare Earth Doped $EuTiO_3$

$Eu^{+2}Ti^{+4}O_3$



Eu 4*f* spins (*S* = 7/2)

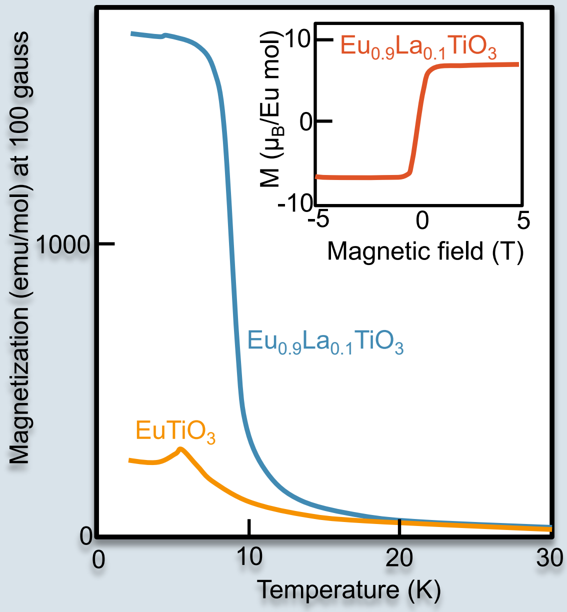
G-type Antiferromagnetic order: T_N = 5.5 K

Cubic at room temperature

Antiferrodistortive transition (I4/mcm)

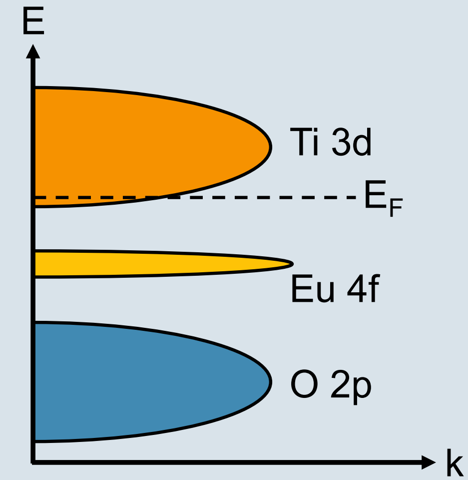
PHYSICAL REVIEW B VOLUME 60, NUMBER 22 1 DECEMBER 1999-II

Transport and magnetic properties of a ferromagnetic metal: $Eu_{1-x}R_xTiO_3$



T. Katsufuji and Y. Tokura, Phys. Rev. B 60, R15021 (1999).

***R*:$EuTiO_3$ (*R* = La, Gd,…)**



Carrier mediated (ferro)magnetism and transport?

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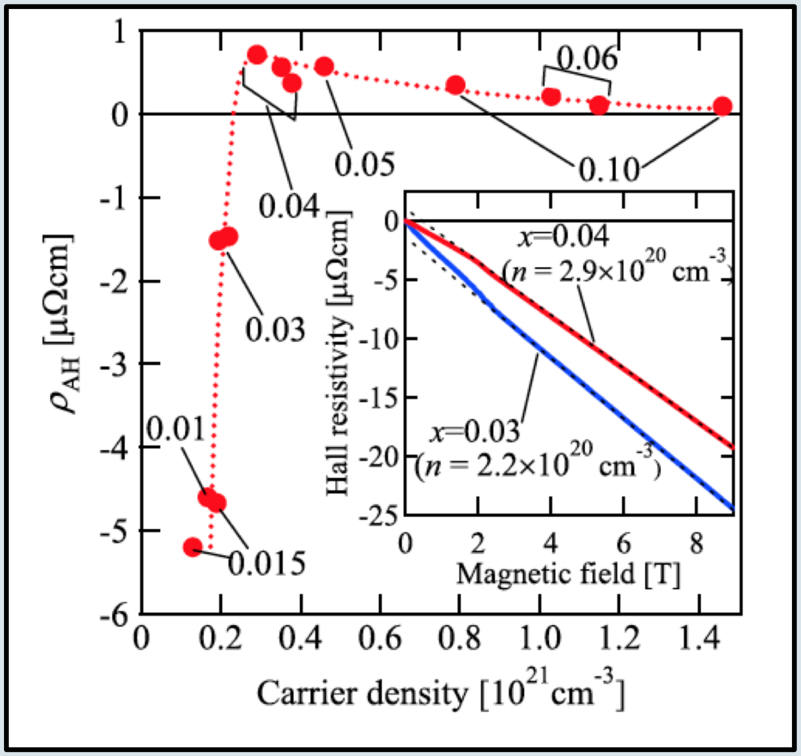
Rare Earth Doped $EuTiO_3$



K. S. Takahashi et al., PRL 103, 057204 (2009).

- $EuTiO_3$: Insulating AFM below 5 K
- $Eu_{1-x}La_xTiO_3$
- Metallic (anti-)ferromagnet

  T. Katsufuji and Y. Tokura, Phys. Rev. B 60, R15021 (1999).
- Sign change in anomalous Hall effect with carrier density
- Suggests intrinsic origin of AHE

Why Rare Earth Doped $EuTiO_3$?

- $Eu_{1-x}R_xTiO_3$ (R = trivalent rare earth ion such as Sm or La)
- Metallic (anti-)ferromagnet T. Katsufuji and Y. Tokura, Phys. Rev. B 60, R15021 (1999).

- Sign change in AHE with carrier density suggests intrinsic, topological origin
  → **Band crossing**
  → Possibly other types of interesting phenomena: changes in the Fermi surface topology by doping, magnetic field, …

- Low carrier densities (compared to typical metals)
  → Low Fermi level
  → Magnetic energy scales can drive transitions

- Low carrier densities → electric field-effect control

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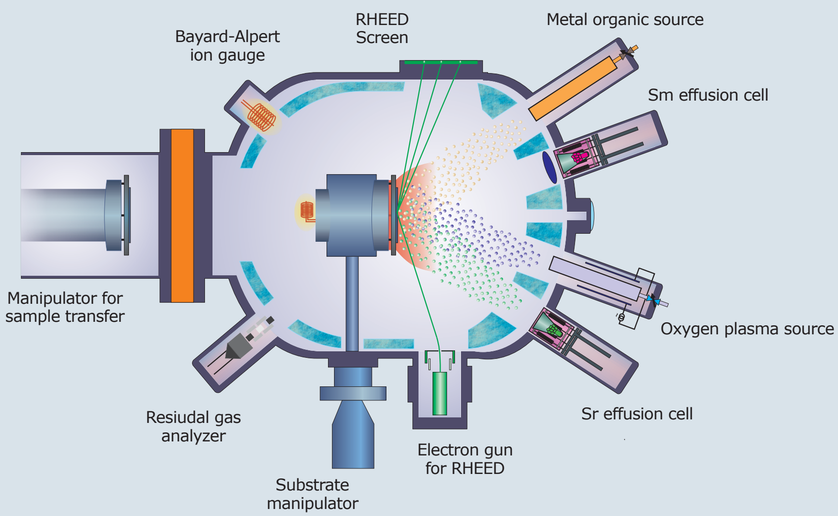
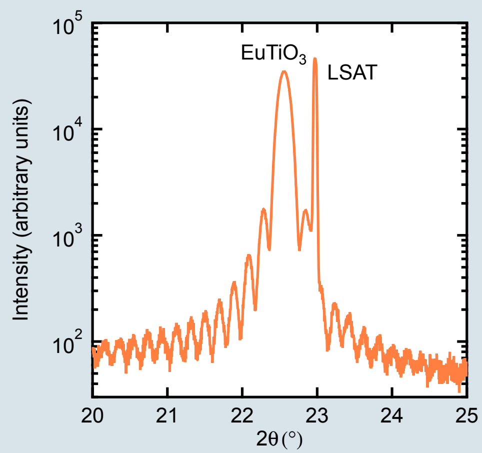
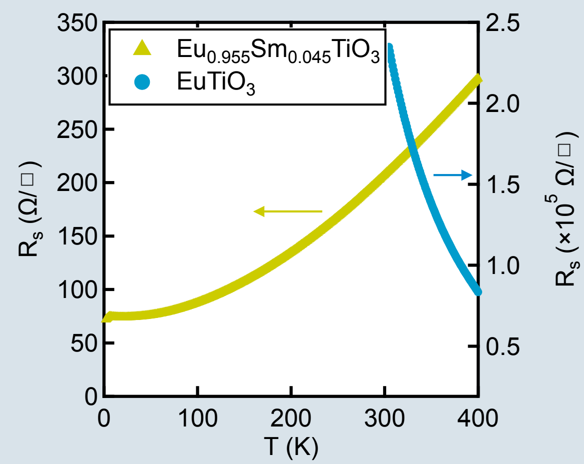
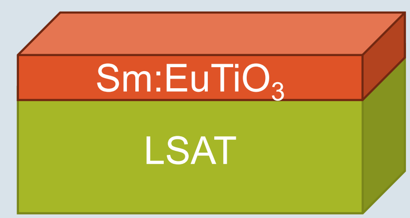
$EuTiO_3$ Films by Molecular Beam Epitaxy









- High-quality epitaxial films using a metal-organic precursor (TTIP) as Ti and oxygen source
- Electron doping with Sm causes films to become metallic
- Vary doping concentration: **1 – 9×10^{20}cm^{-3}**

K. Ahadi, L. Galletti, and S. Stemmer, Appl. Phys. Lett. 111, 172403 (2017).

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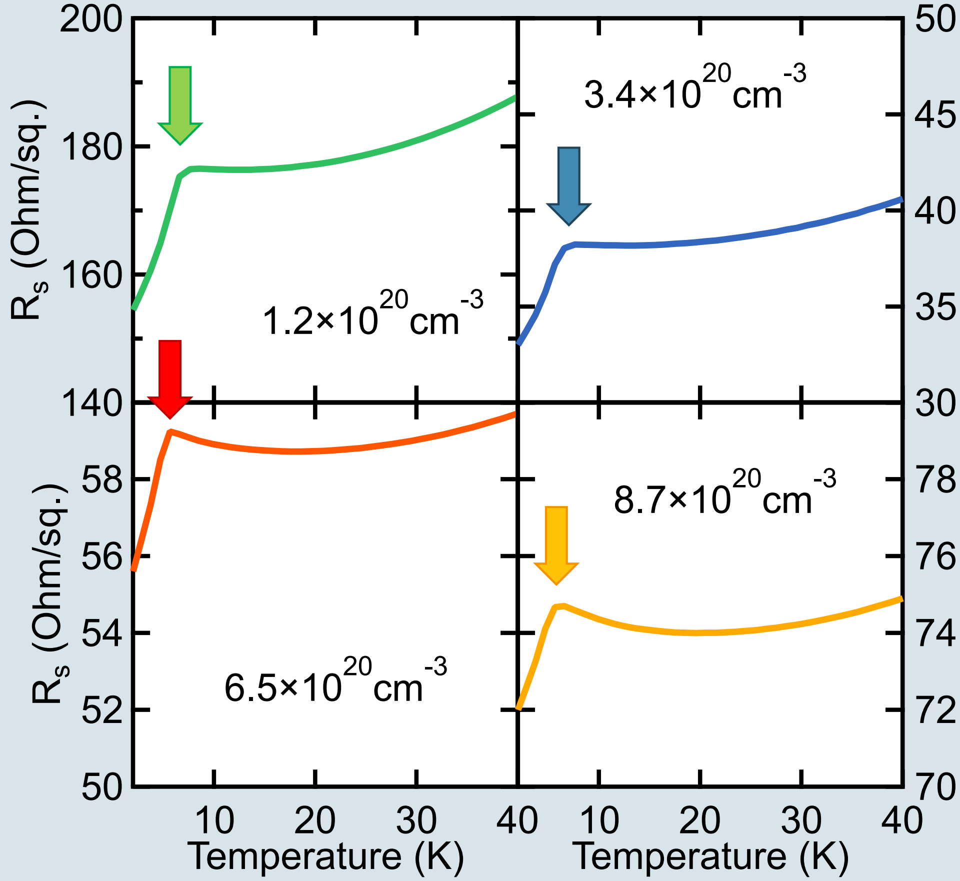
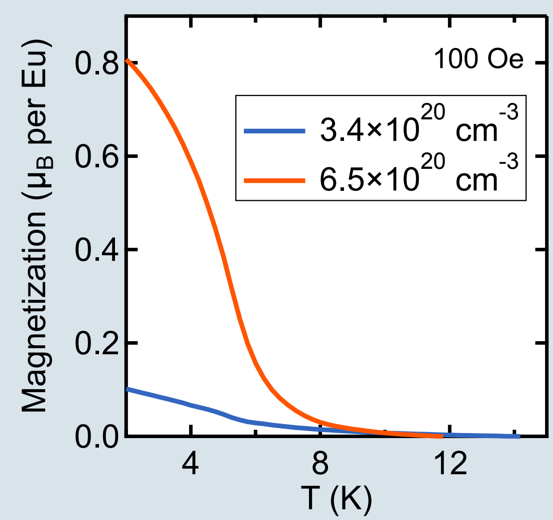
Carrier Mediated Magnetism?



- Resistance anomaly at the magnetic ordering temperature
- Magnetic ordering temperature barely changes with doping concentration
- Similar to T_N of undoped $EuTiO_3$
- Change in magnetic order with doping?



K. Ahadi, et al., APL Materials 6, 056105 (2018).

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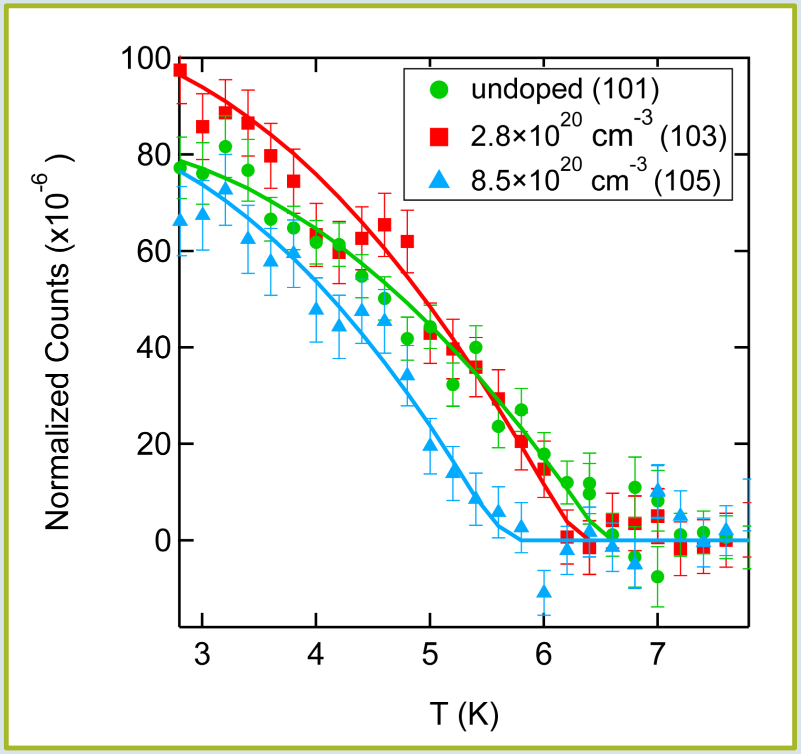
Carrier Mediated Magnetism?



- Neutron diffraction measurements as a function of doping
- Magnetic Bragg peaks at tetragonal [odd, 0, odd] (pseudocubic [½ ½ ½]-type) observed in all samples below T_N
- Persistent antiferromagnetic order with carrier substitution
- Only small change in T_N with doping

K. Ahadi, et al., APL Materials 6, 056105 (2018).

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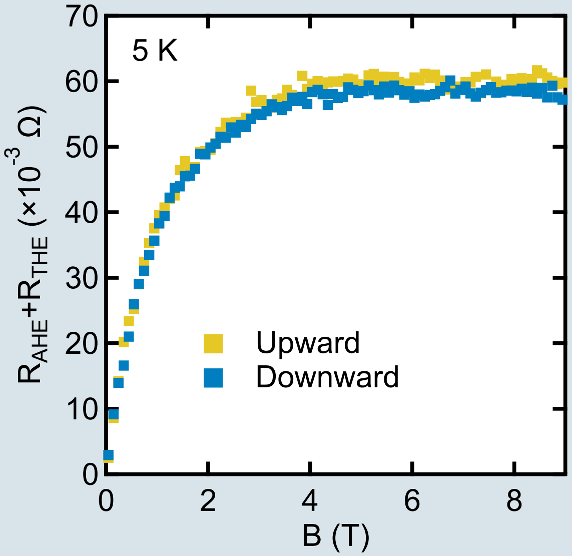
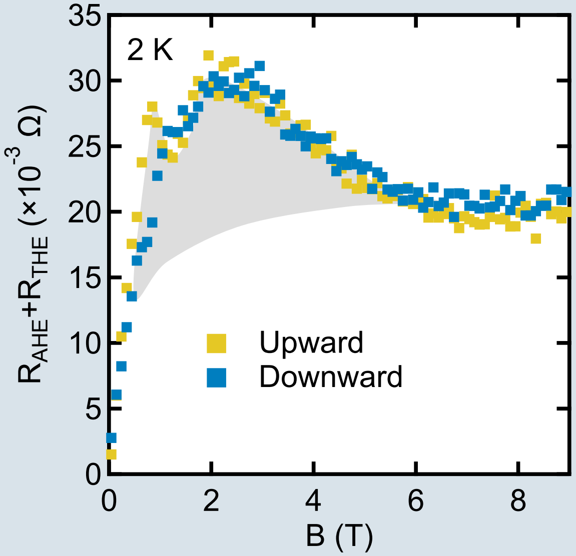
Spontaneous Hall Effects

$R_{xy} = R_0B + R_{AHE} + R_{THE}$

n = 9×10^{20}cm^{-3}

AHE



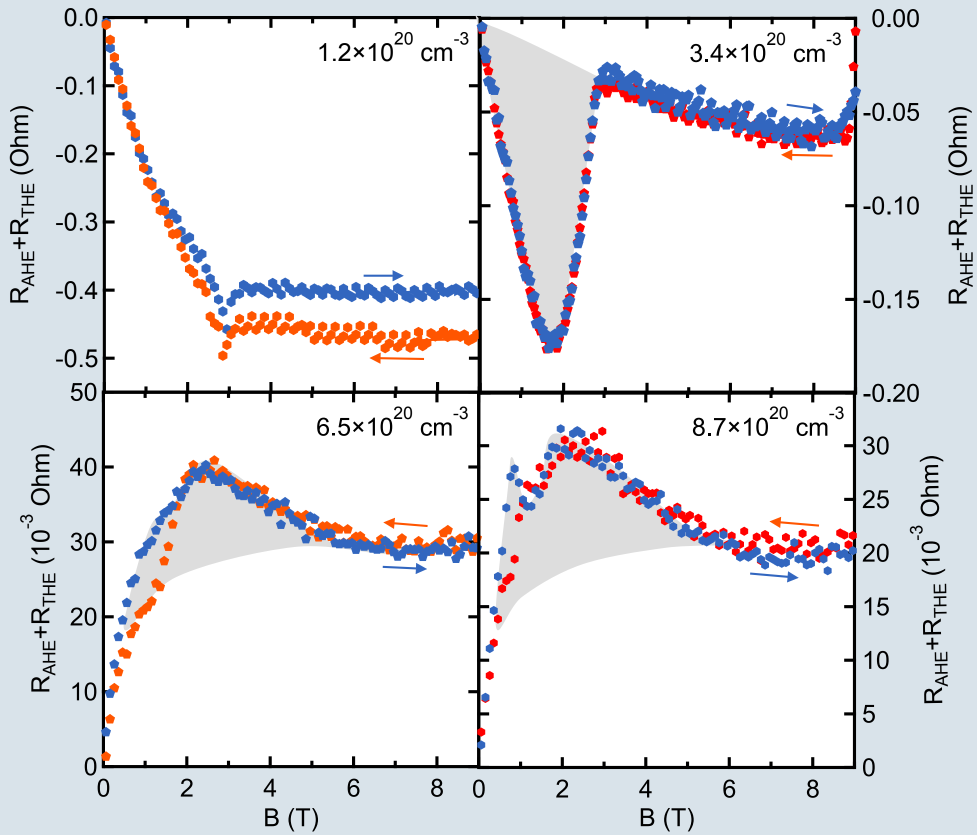


AHE+THE

- Below ~ 30 K: Anomalous Hall effect (AHE)
- Below 5 K: Additional non-monotonic contribution

Ahadi et al., Appl. Phys. Lett. 111, 172403 (2017). UCSB

Spontaneous Hall Effects



K. Ahadi, et al., APL Materials 6, 056105 (2018).

- Different carrier densities
- Both spontaneous Hall effects at all carrier densities
- Despite small net magnetization
- Sign change in BOTH effects at ~ 4×10^{20} cm^{-3}
- **Change in both reciprocal (and real space) Berry curvatures**
- Reciprocal space: Weyl points?

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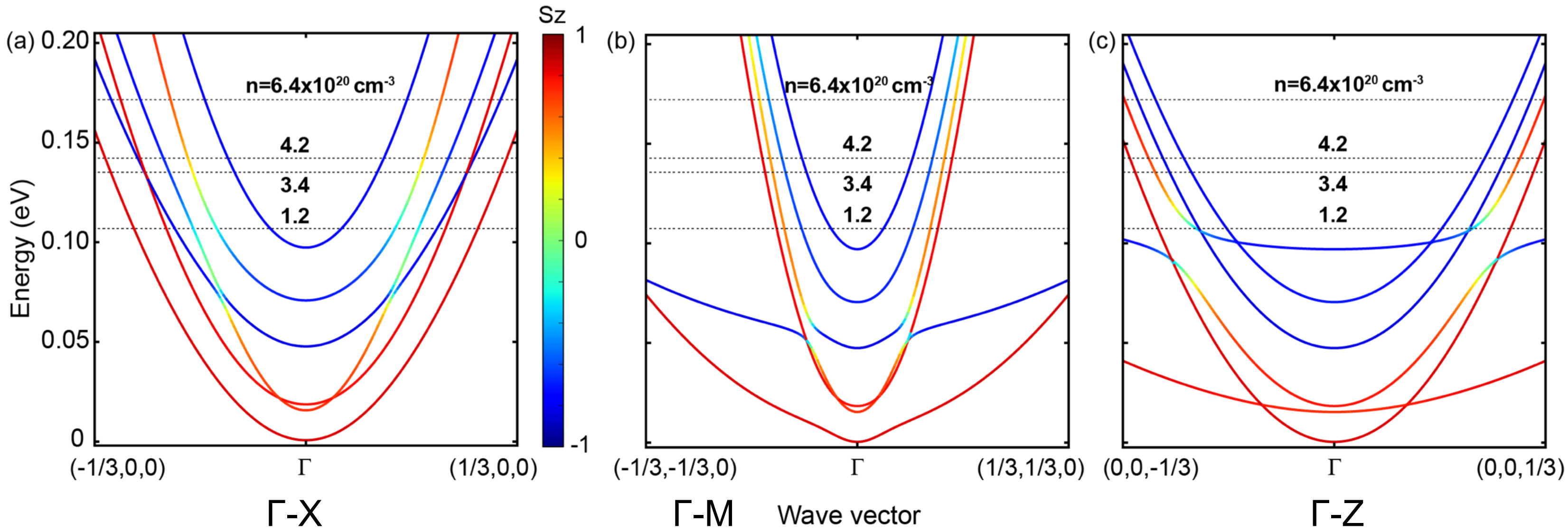
Anomalous and Topological Hall Effects



- DFT calculations (Janotti group)
- Ti-3d conduction band with spin-up crosses another Ti-3d conduction-band with spin-down along the Γ-X (in-plane) direction, in a spin-momentum locked configuration at the Fermi level for **~ 4×10^{20} cm^{-3}**
- **Source of reciprocal space Berry curvature**

K. Ahadi, et al., APL Materials 6, 056105 (2018).

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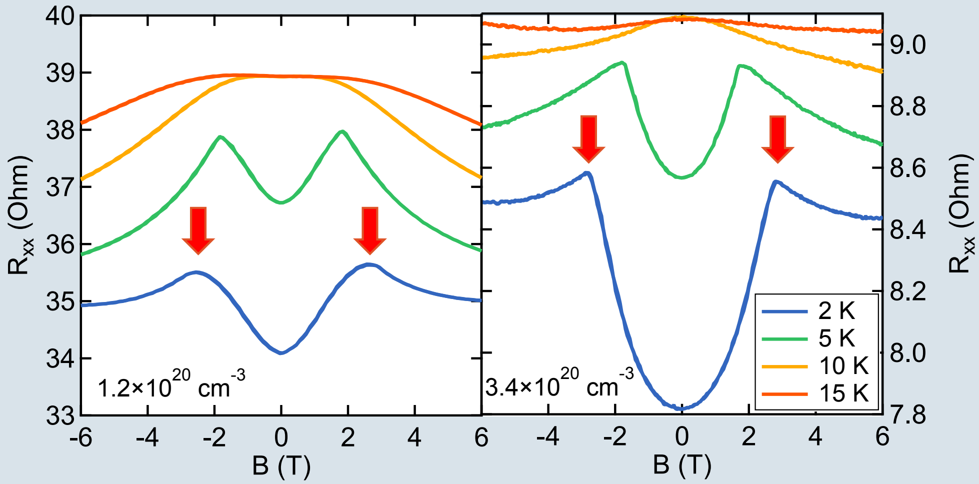
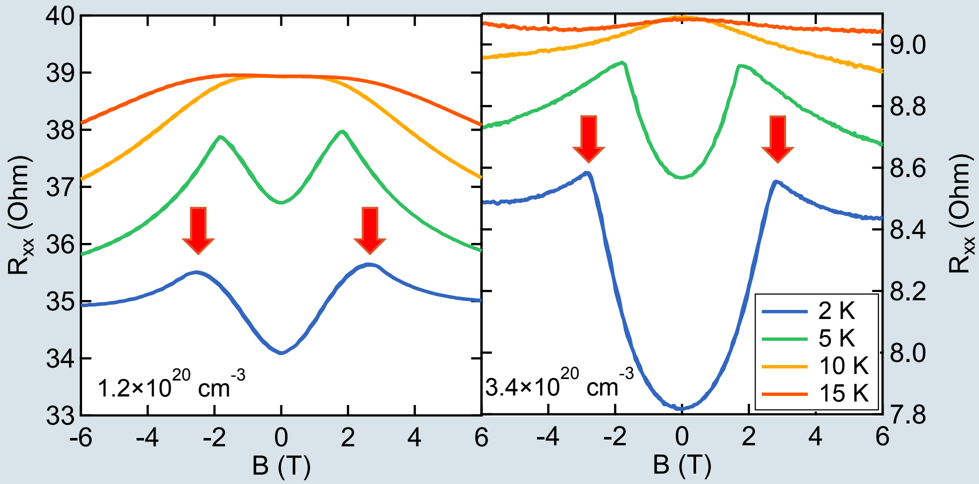
Magnetoresistance

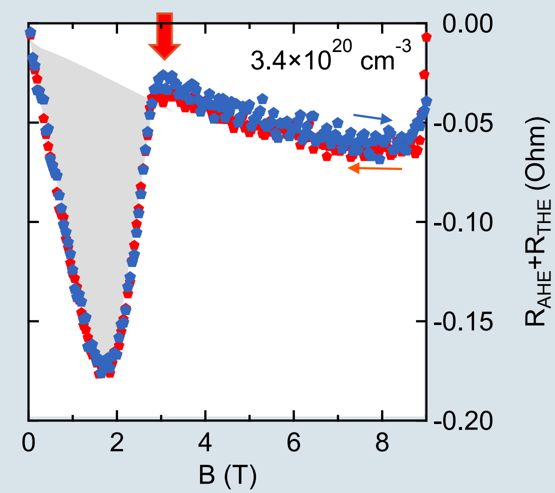


- Low doping
- Low-magnetization state
- Metamagnetic transition: non-collinear AFM → FM

Magnetoresistance



- Low doping
- Low-magnetization state
- Abrupt phase transition with field
- Metamagnetic transition: non-collinear AFM → ?



THE vanishes near at the metamagnetic transition

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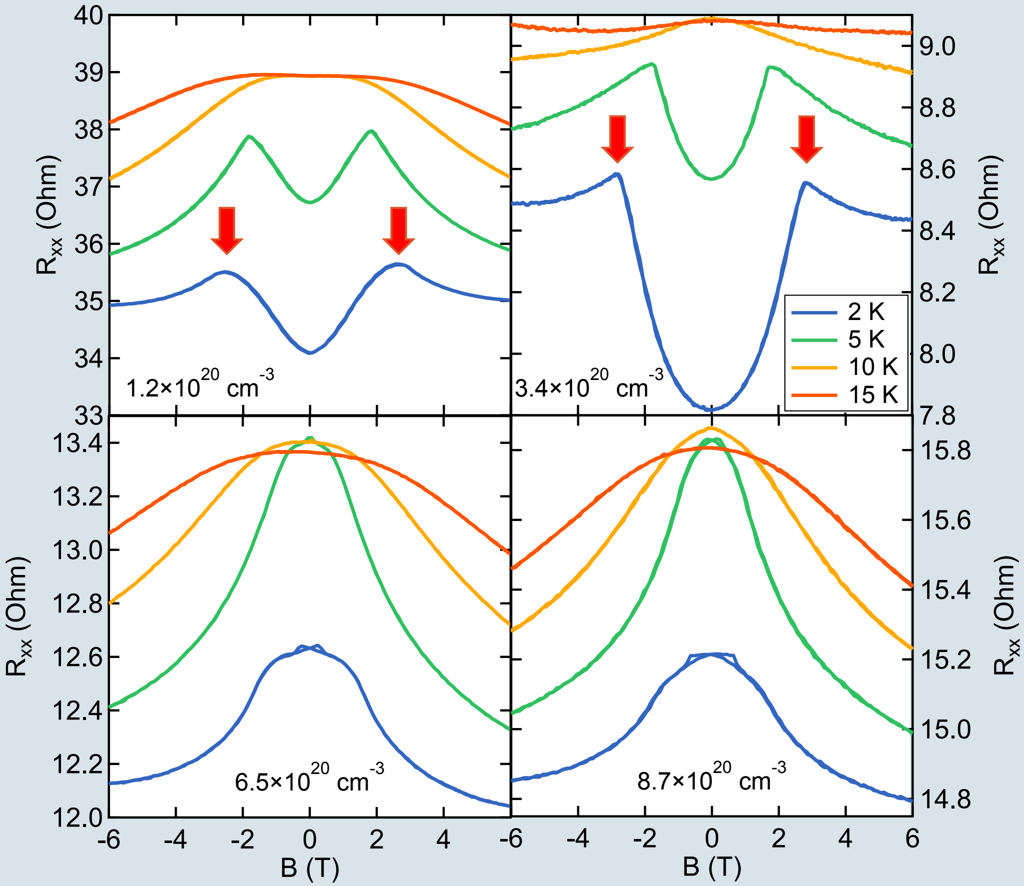
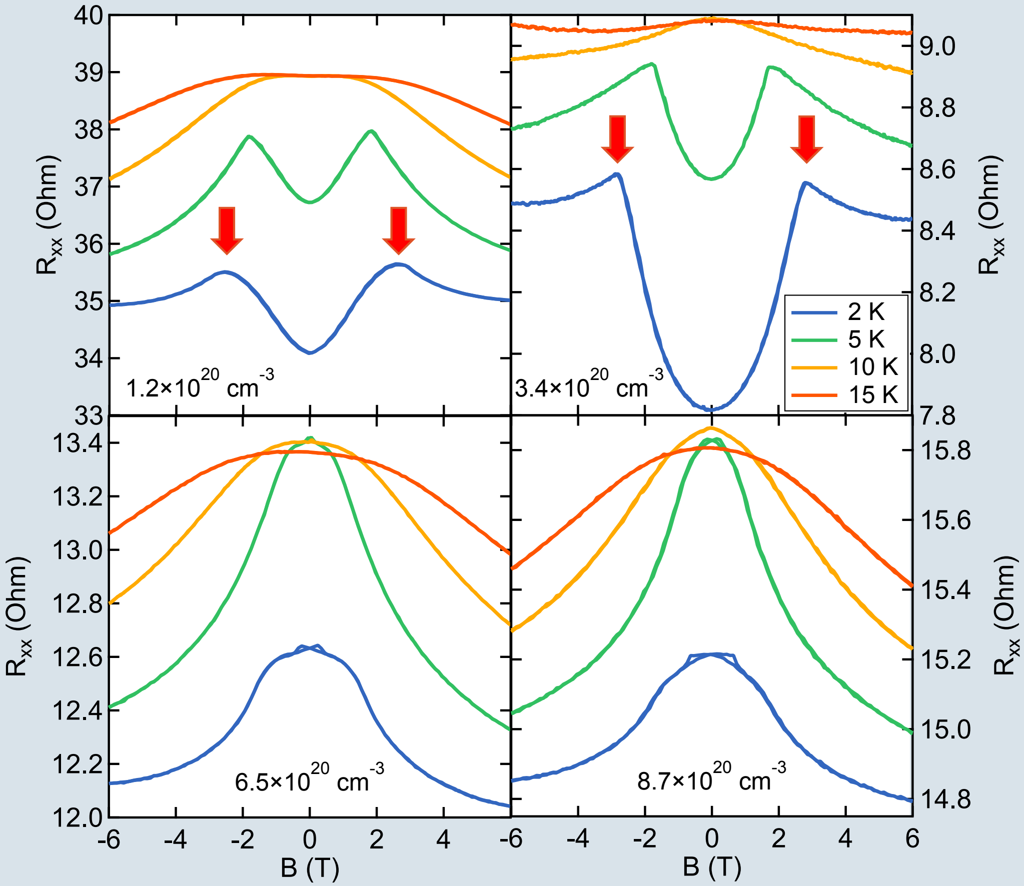
Magnetoresistance



- High doping
- High-magnetization state
- Negative MR: only a gradual change from a non-collinear spin structure to a more collinear one

Magnetoresistance



- Low doping
- Low-magnetization state
- Abrupt phase transition with field
- Metamagnetic transition: non-collinear AFM → ?

Presence/absence of phase transition coincides with sign changes in Hall effects, Berry curvature, and Fermi level crossing the Weyl point.

- High doping
- High-magnetization state
- Negative MR: only a gradual change from a non-collinear spin structure to a more collinear one

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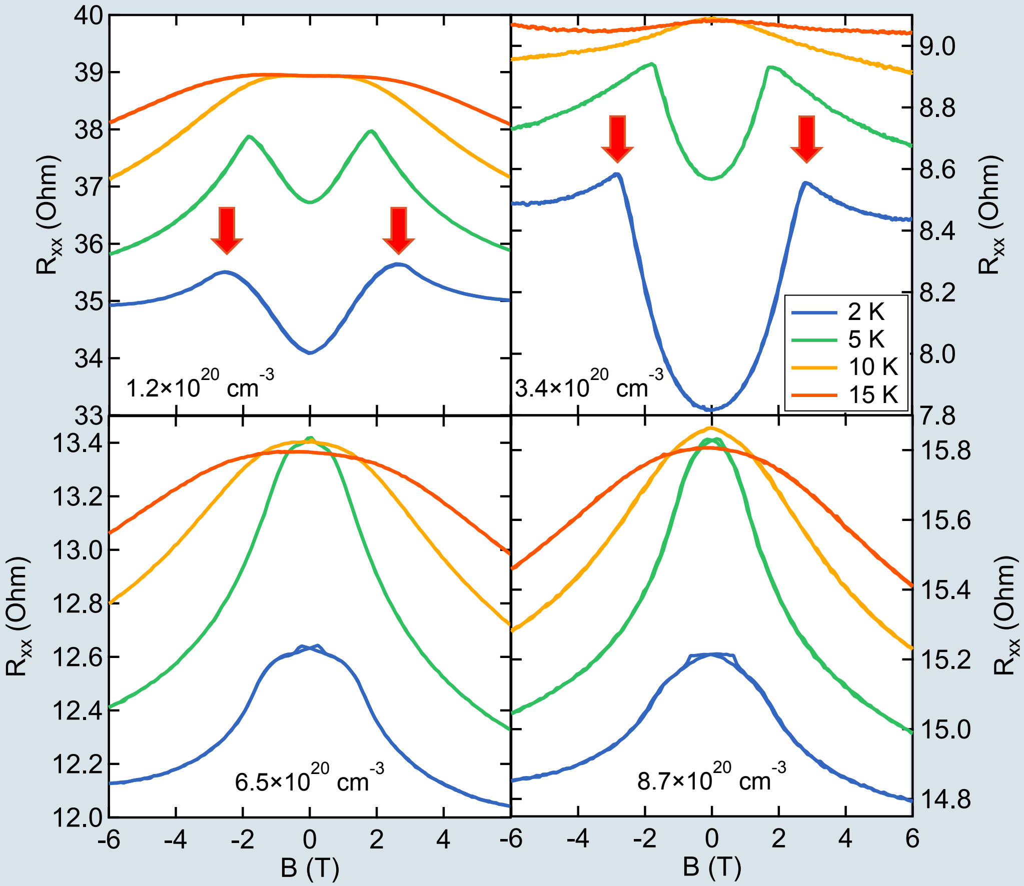
Magnetoresistance



- Low doping
- Low-magnetization state
- Abrupt phase transition with field
- Metamagnetic transition: non-collinear AFM → ?

Strong connection between magnetism and Fermi surface topology.

- High doping
- High-magnetization state
- Negative MR: only a gradual change from a non-collinear spin structure to a more collinear one

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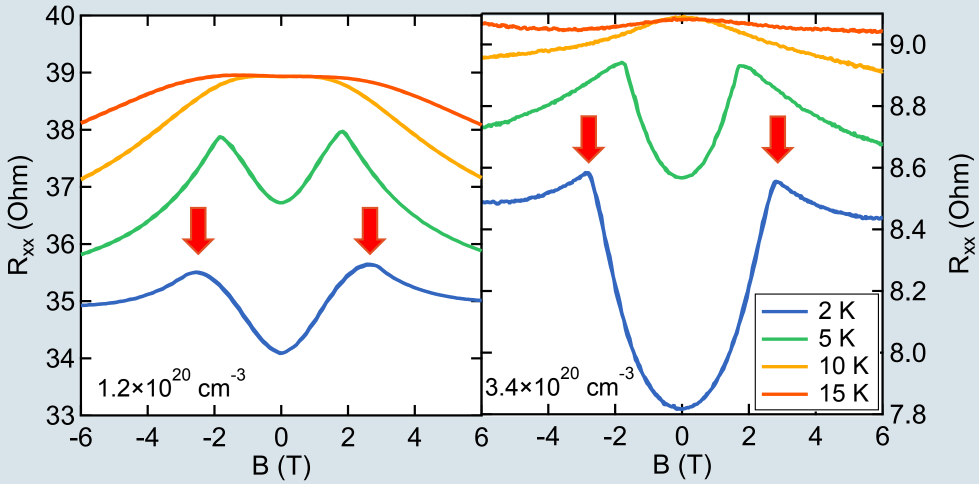
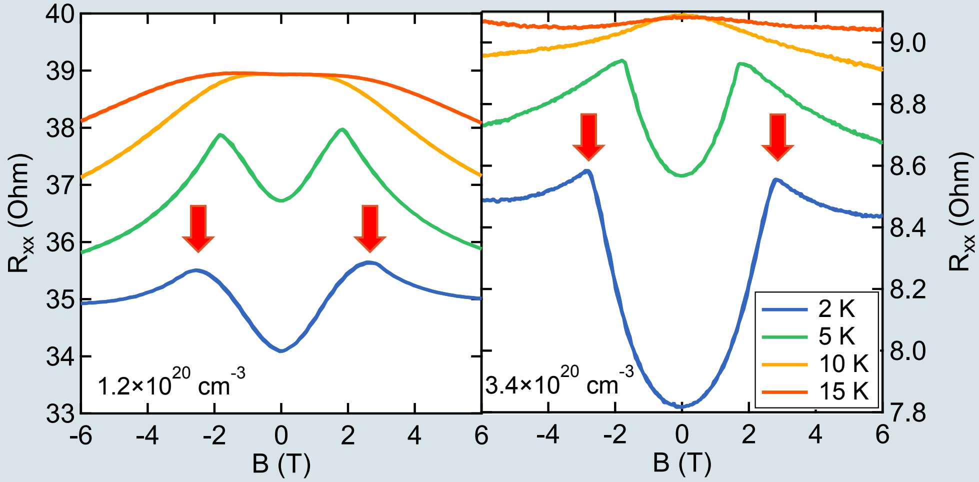
Metamagnetic Transition

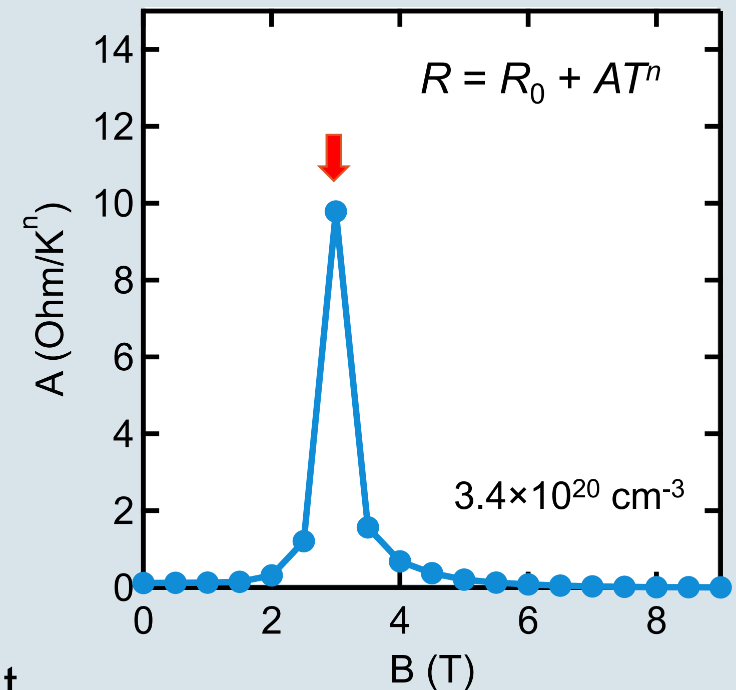


- Metamagnetic transitions are often associated with singularities in the density of states and Zeeman-splitting induced change in Fermi surface topology (Lifshitz transitions)
- Presence of Weyl points suggest another possibility: as the magnetic field reorients the spins, it modifies the band topology and thereby magnetotransport

K. Ahadi, et al., APL Materials 6, 056105 (2018).

Metamagnetic Transition



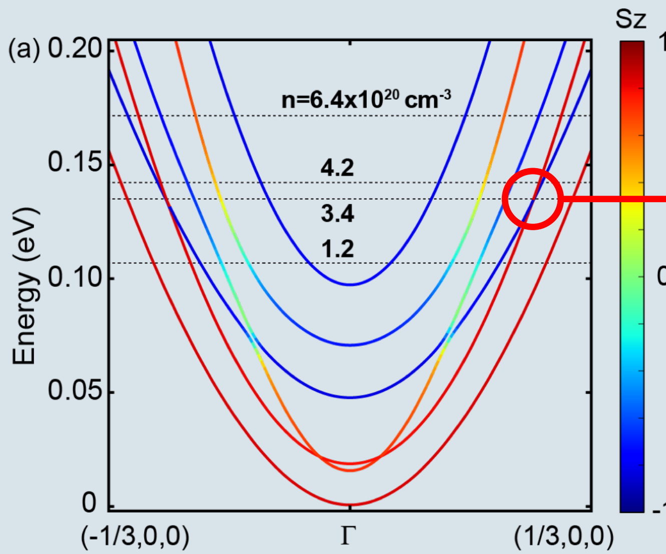


$$\ln\left(\frac{dR(T)}{dT}\right) = \ln(nA) - (n-1)\,ln(T)$$

- Peak in A parameter (mass enhancement) near metamagnetic transition suggests magnetic field controlled quantum critical point
- Magnetic field controlled Fermi surface fluctuations/instability

K. Ahadi, et al., APL Materials 6, 056105 (2018). UCSB

Magnetism and Electronic Structure



Doping density above critical E_F

Doping density below critical E_F

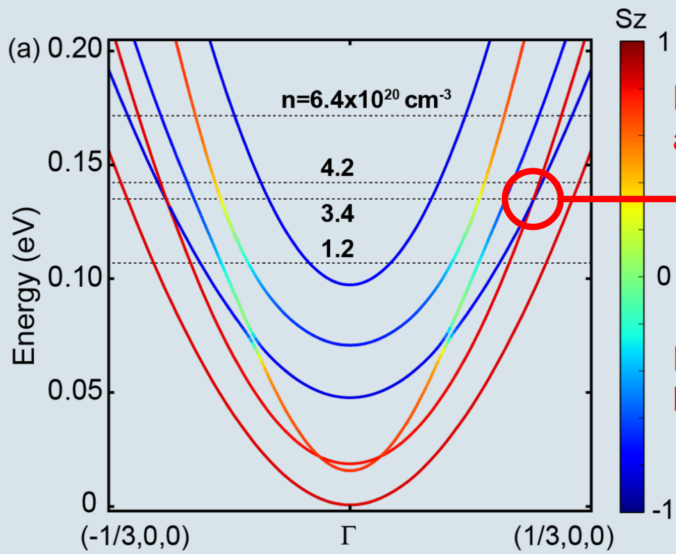
Magnetic field causes gradual change to more collinear spin structure

Critical Fermi level → Sign change in spontaneous Hall effects

- Magnetic field induced change in Fermi surface
- Change in spin texture driving FS change
- Quantum critical
- Non-monotonous spontaneous Hall effect vanishes at the transition

K. Ahadi, et al., APL Materials 6, 056105 (2018).

Magnetism and Electronic Structure



Doping density above critical E_F

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What happens when we modify the electronic band structure, i.e. 3D → quasi-2D?

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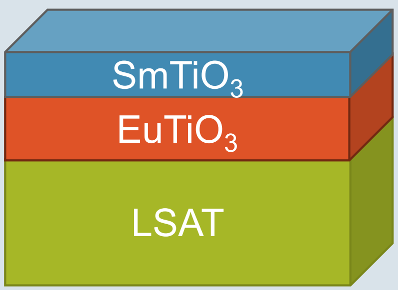
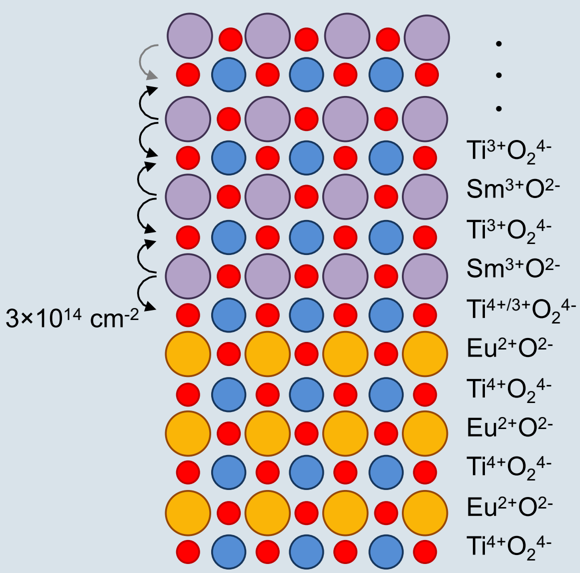
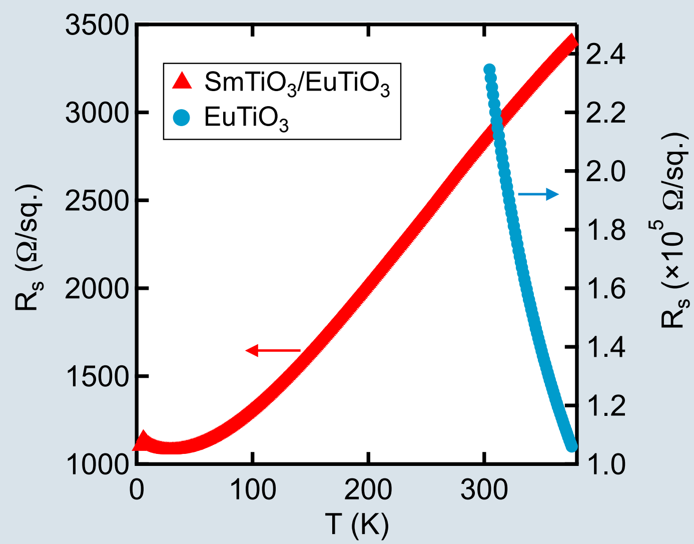
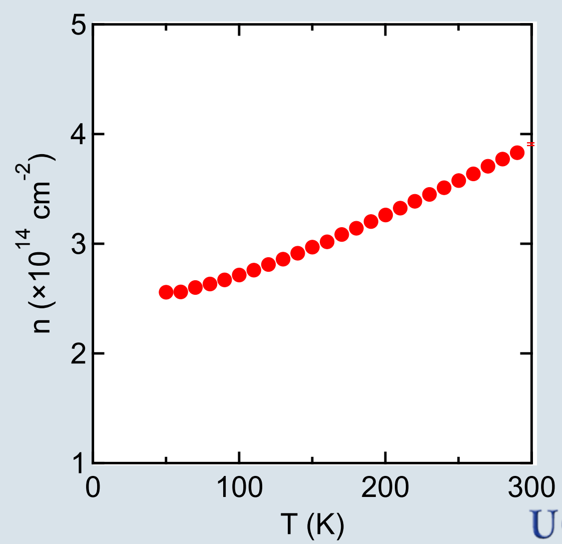
$SmTiO_3/EuTiO_3$ Interfaces

- $SmTiO_3/EuTiO_3$ interfaces: quasi-two dimensional electron system **in the $EuTiO_3$**, similar to $SmTiO_3/SrTiO_3$ interfaces
- Full mobile charge density expected from polar discontinuity: 3.4×10^{14} cm^{-2}
- Charge spreads into the $EuTiO_3$ layer: “quasi-2D”









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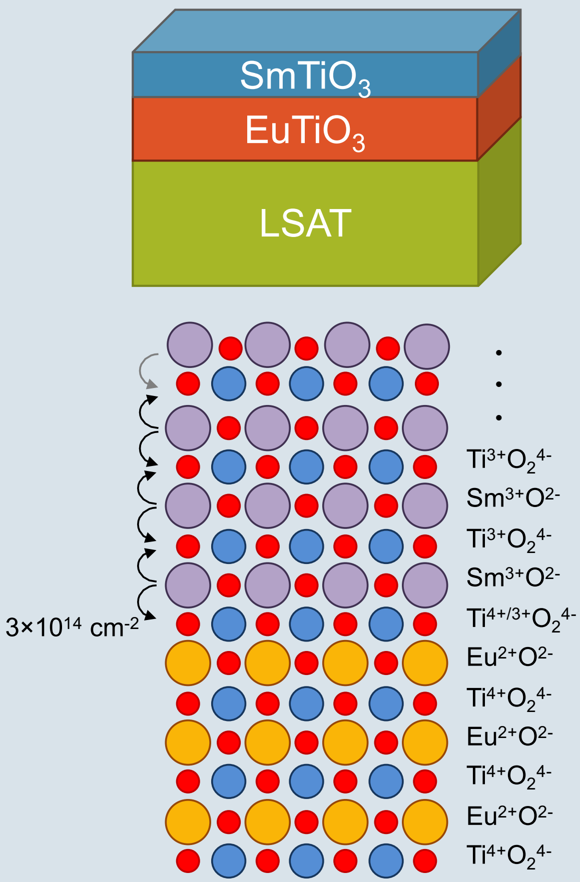
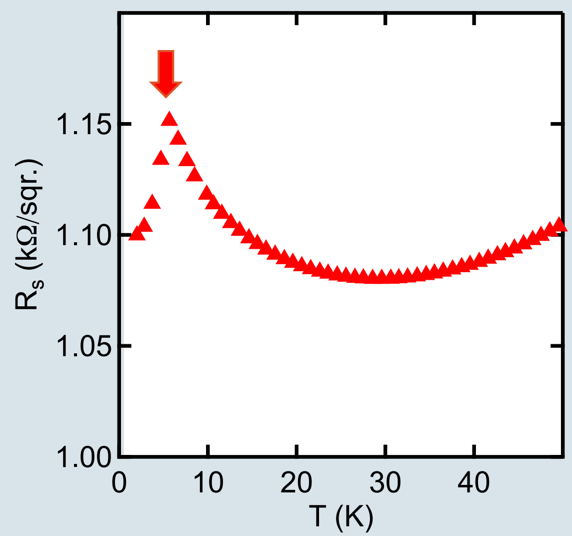
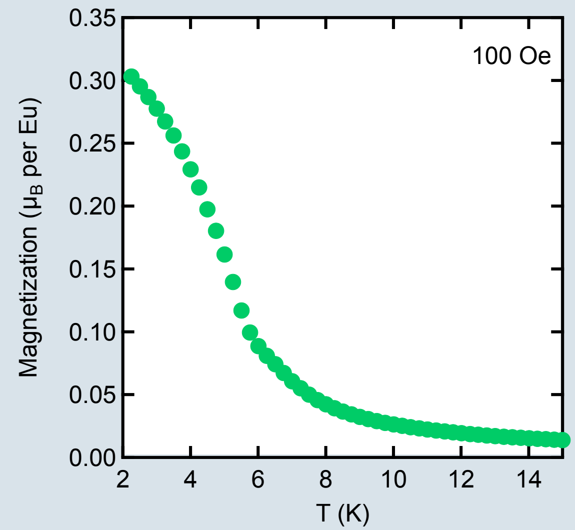
$SmTiO_3/EuTiO_3$ Interfaces



- Similar magnetic transition temperature as bulk $EuTiO_3$
- Similar magnetization as highly doped $EuTiO_3$ films
- Mobile charge resides in the $EuTiO_3$





K. Ahadi, H. Kim, and S. Stemmer, APL Mater. 6, 056102 (2018)

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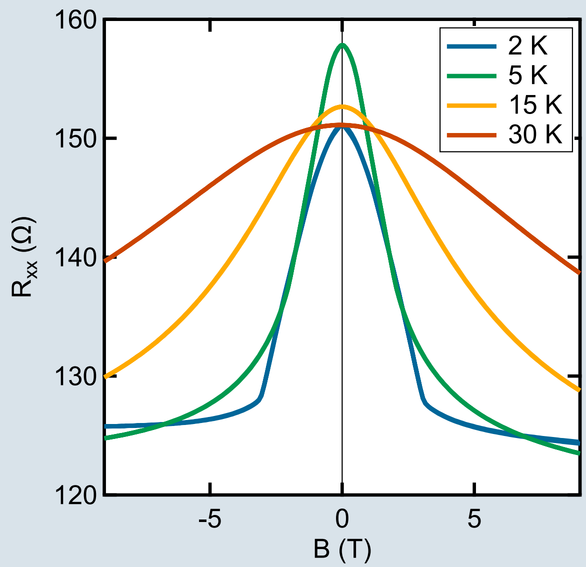
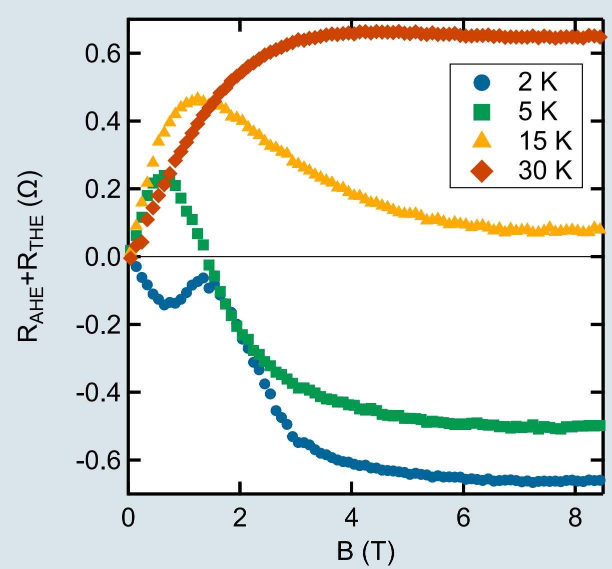
$SmTiO_3/EuTiO_3$ Interfaces

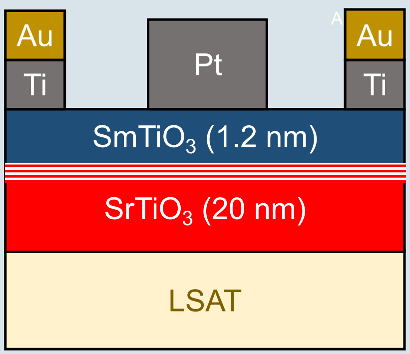
K. Ahadi, H. Kim, and S. Stemmer, APL Mater. 6, 056102 (2018)

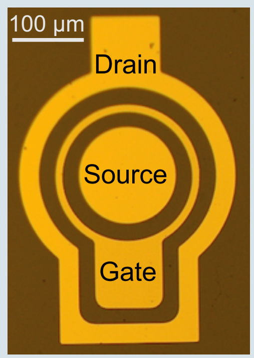


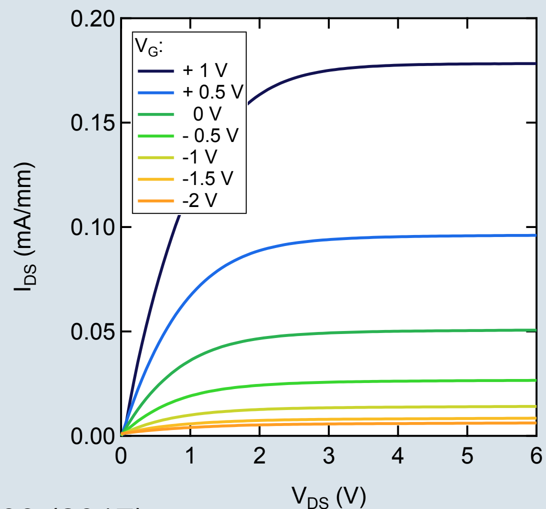


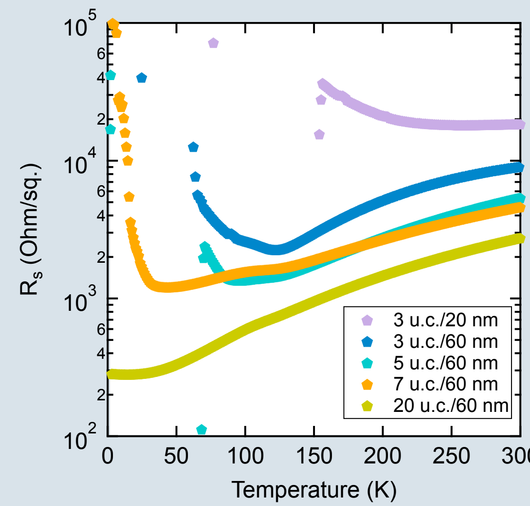
- AHE changes sign upon lowering the temperature → Fermi level closer to points with change in Berry curvature
- THE appears at higher temperatures than in uniformly doped $EuTiO_3$, but does not change sign
- Interface stabilizes non-collinear spin texture to higher temperatures
- Results consistent with a picture of AHE as a signature of reciprocal space Berry curvature and THE as being due to real space Berry curvature

Outlook









K. Ahadi, et al., Appl. Phys. Lett. 110, 062104 (2017).

K. Ahadi and S. Stemmer, Phys. Rev. Lett. 118, 236803 (2017).

- ❑ Interfaces with thin $SmTiO_3$ gate dielectrics can be gated (fully depleted)
- ❑ Novel metal-insulator transition: correlation effects?
- ❑ Electron correlations and topology?

Summary

- Carrier density tuning of spontaneous Hall effects in doped $EuTiO_3$
- AHE despite small net magnetization
- Sign changes in spontaneous Hall effects with carrier density
- Carrier density control of magnetic field induced transition of the FS and quantum critical points
- Common origin: band topology (crossings): magnetic topological “semimetal”
- Results raise interesting questions as to connection between real space magnetic topology and reciprocal space topology
- **Theory needed**
- Interesting system for electric field gating

UCSB

Thank you!